Warsaw, 30.05.2014

A SUMMARY OF SCIENTIFIC ACHIEVEMENTS

1. NAME AND SURNAME

Ryszard Broda
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2. GRADUATION DIPLOMAS, ACADEMIC / ARTISTIC DEGREES – UNIVERSITY, PLACE AND YEAR OF OBTAINING:

1975	Master of Science in Physics, Warsaw University
1994	Doctor of Physics, Institute of Atomic Energy, Świerk, title: "*Numerical method of activity determination by the liquid scintillation technique*"

3. EMPLOYMENT WITH SCIENTIFIC / ARTISTIC INSTITUTES:

1975 - 82	Junior Researcher, Researcher, Metrological Laboratory, Centre for Production and Distribution of Isotopes, Institute of Nuclear Research, Świerk
1983 - 94	Senior Researcher, Radioisotope Centre (RC, former: Reactors and Radioisotopes Production Centre), Institute of Atomic Energy, Świerk
1994 -	Assistant Professor, RC, Świerk
1996 - 2002	Head, Metrological Laboratory of Radioactive Materials, RC , Świerk
2002 - 2004	Research & Development Unit, RC , Świerk
2005 - 2006	Head, Laboratory of Radioactivity Standards, RC , Świerk
2006 -	Laboratory of Radioactivity Standards (LWR), Radioisotope Centre POLATOM, Institute of Atomic Energy, (from 2011: National Centre for Nuclear Research Radioisotope Centre POLATOM), Świerk

4. ACHIEVEMENTS RESULTING FROM ART. 16 SECTION 2 OF THE ACT OF 14 MARCH 2003 ON ACADEMIC DEGREES AND TITLE AND DEGREES AND TITLE IN THE ARTS (JOURNAL OF LAWS NO 65, ITEM 595 AS AMENDED):

a) author/authors (the author's contribution in %), title/titles of publications, year of issue, publisher's name,

As the scientific achievement to be indicated under the above-given law, I have chosen a monothematic cycle of 9 publications on application the detection efficiency of liquid-scintillation counters calculation to determine the radionuclides activity in the triple-to-double coincidence ratio (TDCR) method. I present these publications below with an indication of the *Impact Factor* of the journal in the year of publication, number of quotations according the Web of Science (WoS) base and percentage contribution based on my own estimation:

	Title of publication	*Impact factor*	Number of quotations acc. to WoS	My contri-bution
[1]	**Broda R.,** Péron M.N., Cassette P., Terlikowska T., Hainos D. Standardization of ^{139}Ce by liquid scintillation counting using the triple to double coincidence ratio method. *Appl. Radiat. Isot.*, 49, (1998) 1035-1040.	0.598	3	50 %
[2]	**Broda R.,** Cassette P., Małetka K., Pochwalski K. A simple computing program for application of the TDCR method to standardization of pure-beta emitters, *Appl. Radiat. Isot.*, 52, (2000) 673-678.	0.716	15	60 %
[3]	Cassette P., **Broda R.,** Hainos D., Terlikowska T. Analysis of detection-efficiency variation techniques for the implementation of the TDCR method in liquid scintillation counting. *Appl. Radiat. Isot.*, 52, (2000) 643-648.	0.716	16	35 %
[4]	**Broda R.,** Małetka K., Terlikowska T., Cassette P. Study of the influence of the LS-cocktail composition for the standardisation of radionuclides using the TDCR model. *Appl. Radiat. Isot.*, 56, (2002) 285-289.	0.768	9	40 %
[5]	**Broda R.**, Jęczmieniowski A. Statistics of the LS-detector in the case of low counting efficiency. *Appl. Radiat.Isot.,* 60, (2004) 453-458.	1.000	4	85 %
[6]	**Broda R**. Some remarks on photons statistics in the LS-counter. *Appl. Radiat. Isot.*, 66, (2008) 1062-1066.	1.114	4	100 %
[7]	**Broda R.** A review of the triple-to-double coincidence ratio (TDCR) method for standardizing radionuclides. *Appl. Radiat.Isot.*, 58, (2003) 585-594.	0.690	17	100 %
[8]	**Broda R.**, Cassette P., Kossert K. Radionuclide metrology using liquid scintillation counting. *Metrologia*, 44, (2007) S36-S52.	1.667	51	30 %

b) discussion of the scientific / artistic goal(s) of the above work / works, the obtained results and description of their likely utilization,

The title of the scientific goal of the above works is as follows:

"The LS-counters detection efficiency in the measurements of radionuclides activity"

The above-given scientific goal is related to the investigatinon into the ways of measuring the radionuclides activity, particularly of pure β-emitters, in the triple-to-double coincidence ratio (TDCR) method using the liquid scintillation counting (LSC). The above-mentioned publications were referred to in the *Handbook of radioactivity analysis* (Ed.: M.F. L'Annunziata, Acad. Press, Elsevier Inc. Amsterdam, Boston, Heidelberg, London, New York, Oxford, Paris, San Diego, San Francisco, Sydney, Tokyo. 2012) which is a unique standard reference source for the metrological community with respect to teaching, research and peaceful applications of radioactive sources.

Introduction

The triple-to-double coincidence ratio (TDCR) method as published in the report of the Institute of Nuclear Research (Pochwalski K., Radoszewski T. *Disintegration rate determination by liquid scintillation counting using the triple to double coincidence ratio TDCR method.* INR Report, 1847/OPiDI/E/A, 1979, Warsaw) was conceived and developed by K. Pochwalski, PhD. Eng. in the OPiDI IBJ in Świerk in 1978. The method is intended for determining radionuclide activity, especially of pure β-emitters such as ^{3}H, ^{14}C or ^{63}Ni and low-energy electron-capture radionuclides, e.g. ^{55}Fe and ^{54}Mn, which are difficult to measure by means of other methods. The consecutive development and improvement of the TDCR method allowed its application also in the measurements of activity of γ-emitters with a complex decay scheme. An important element of the correct application of the TDCR method is the calculation of the LS-counters detection efficiency.

The TDCR method is the only absolute method of determination of pure β-emitters activity in the LSC technique. The solution of a given radionuclide is mixed with a liquid scintillation cocktail being a mixture of a primary scintillator (solute), an organic solvent, a surfactant, that ensures miscibility of the radioactive aqueous sample with the organic solution, and the secondary scintillator (wave-length shifter) matching the scintillator emission spectrum to the photomultiplier (PMT) photocathode absorption spectrum. The charged particles emitted following the radionuclide decay lose the energy which is absorbed in the solvent and then propagated to the scintillator. The emitted scintillation photons are registered by PMTs of the detector. The scintillation counter has three PMTs (the triple counter) that register scintillation pulses in the channels of different coincidences. The activity of the radionuclide is determined by extrapolating the counting points to the full detection efficiency of the detector as a function of a specific experimental parameter of the TDCR method, being the ratio of triple coincidence count rates to the logical sum of the double coincidences count rates. K. Pochwalski proposed to use an ordinary linear extrapolation. I replaced this relatively simple but approximate way of determining the activity with a theoretical calculation of the efficiency of a triple counter based on the description of the physical phenomena occurring in the counters and use of a nonlinear extrapolation function. The function values were determined through numerical calculations based on the theory of radioactive decay and appearance of nuclear radiation, its interaction with the matter and transfer of the energy exciting the solvent molecules. The ionization quenching was taken into account. This phenomenon reduces the energy propagation and the number of emitted scintillation photons. The ionization quenching is produced by a high ionization density along the charged particle path in the scintillator. Subsequently, I incorporated the collection of the scintillation photons by PMTs, emission of the photoelectrons, PMTs asymmetry and creation various pulses on the detector output in the calculation. I continued to improve the code for computing detection efficiency which considerably reduced the uncertainty concerning the radionuclides activity measured using the TDCR method and extended the application of the method to radionuclides with various decay schemes. An important element of the correct application of the TDCR method is the calculation of the LS-counters detection efficiency based on the description of the physical phenomena occurring in the counters.

In 1982, upon Pochwalski's initiative and in his presence, the TDCR method was adopted by the Laboratoire de Metrologie des Rayonnements Ionisants (LMRI), Saclay, France. It is now applied globally by all leading laboratories of radionuclide metrology where the TDCR detectors were constructed. The TDCR method became one of the fundamental and efficient tools of radionuclide metrology for β-emitters and low-energy electron-capture radionuclides (EC-nuclides) standardization. The method was incorporated into the International Reference System (SIR) for measuring the activity of β-emitters established at the Bureau International des Poids et Mesures (BIPM) in Sèvres and continues to be developed by the Laboratory of Radioactivity Standards at the National Centre for Nuclear Research Radioisotope Centre POLATOM in Świerk. The routine activity measurements using the TDCR method and determination of the concentration of radioactive solutions are of fundamental importance for the production and control of standard sources applied in science, industry, nuclear medicine and environmental protection.

Description of the Problem

a) Routine measurements of pure β-emitters and low-energy EC-nuclides, which were difficult to standardize by means of the existed radionuclide metrology methods, should be performed using the absolute TDCR method in the triple LS-counter, developed by K. Pochwalski, PhD. Eng. in 1978.

b) The developed model of calculation of the detection efficiency based on the description of physical phenomena occurring in the LS-counters became inherent in the TDCR method but it should be verified based on the analysis of the measurements of activity of various radionuclides in the triple counter.

c) The results of the detection efficiency calculation for low-energy radionuclides were different from the experimental results.

d) It was necessary to expand the LS-counter detection efficiency calculation model for the γ-emitters.

e) The TDCR method together with the LS-counter detection efficiency calculation model have been successively implemented over the last 20 years in all leading radionuclide metrology laboratories around the world.

The elaborated TDCR method appeared to be a very good and indispensable tool for measuring the activity of pure β-emitters, better than any other method used earlier. The method could have been one of the essential methods of the radionuclide metrology in the world. No standard was needed for determination of the measured radionuclide activity as the extrapolation of the experimental points in a function of the specific experimental parameter of the TDCR method, being the ratio of two kinds of coincidence pulses, sufficed. The experimental points were obtained with various LS-counter detection efficiencies. It appeared however, that an ordinary linear extrapolation did not produce results at the desired accuracy level. It was necessary to use a nonlinear extrapolation function based on the model of calculation of the LS-counter detection efficiency I presented in my doctor's thesis. Independence of the result of the detection efficiency was assumed as a criterion of the counting correctness. It was observed that the results of the detection efficiency calculation for low-energy radionuclides were different from the experimental results. During the measurement of the low-energy pure β-emitters, there is always a question of what part of the total number of disintegrations in time unit is not registered considering a limited detection efficiency of a counter with defined noise discriminator level. The question concerns the difference between the theoretical shape of the beta spectrum and the registered spectrum. Without a good understanding of the physical basis of the TDCR method an appropriate evaluation of the measurement results and implementation of the method in the routine activity measurements of radionuclides was not possible. I undertook to solve the problem of the application the model of the LS-counter detection efficiency calculation in the measurements using the TDCR method. I recognized that the initiation of the TDCR method will be of a substantial significance for the standardization of the radioactive sources and solutions of pure β-emitters applied in many fields. I adapted the existed calculation model for γ-emitters [1]. I proved the equivalence of various quenching methods and the invariability of the ionization quenching parameter value [3], [4]. By applying the inductive method, I modified the initial model and put forth a hypothesis that the model of calculation of the LS-counter detection efficiency for low-energy radionuclides ^{3}H and ^{55}Fe needs the Polya distribution for description photoelectrons or PMT-collected photons statistics [5]. I supported the hypothesis with the evidence obtained from the measurements with the light emitting diode (LED) simulating the scintillation source and calculation the approximate shape of the PMT-collected photons statistical distribution [6]. All changes in the detection efficiency calculation model I verified through the measurements of various radionuclide standard sources activity based on a defined criterion of the counting correctness.

Satisfactory results of the test measurements in the triple counter [Zimmerman et al., 2004; Broda et al., 2012]and measurements performed within the framework of international comparisons [Razdolescu et al., 2006; Zimmerman et al., 2010; Makepeace et al., 1998; Cassette et al., 1998] showed that the developed model allows to determine the triple counter detection efficiency with a good approximation and can be applied in routine radionuclide activity measurements by means of the TDCR method. The implementation of the TDCR method in various laboratories stimulated subsequent investigations into the field of the liquid scintillation technique [7].

Description of the Scientific Goal

1. **R. Broda,** M. N. Péron, P. Cassette, T. Terlikowska, D. Hainos. Standardization of ^{139}Ce by liquid scintillation counting using the triple to double coincidence ratio method, *Appl. Radiat. Isot.*, 49, (1998) 1035-1040.

The goal of the above mentioned paper was to verify if the existing model of the pure β-emitters counting efficiency calculation [2, 3] can be also applied to the standardization of γ-emitting radionuclides by means of the TDCR method. The work was performed within the scope of the French-Polish research and technological cooperation, i.e. Joint Project No 194 (212), between the Metrological Laboratory of Radioactive Materials, RC, Świerk and the Laboratoire Primaire des Rayonnements Ionisants (LPRI, former LMRI), Saclay, France. We chose ^{139}Ce, used as a component of the multi-gamma calibration sources, that disintegrates through electron capture following an internal converted gamma transition for measurements.

My contribution to the joint work was an analysis of the ^{139}Ce decay scheme including the emission from the K, L and M shells in the ^{139}La atom and the development of a new algorithm for computing the counting efficiency. I determined the probability of each of the 22 possible atomic rearrangements ways following the electron capture and each of the 24 arrangements following the internal conversion as well as the energies of all the created electrons. I also determined the probability of interaction between the emitted γ and X photons and the liquid scintillator, the probability of the photoelectric or the Compton effect and the spectrum of Compton electrons. Considering the ionization quenching function, I calculated the full scintillation spectrum of ^{139}Ce and the counting efficiency of ^{139}Ce as a function of a specific experimental parameter of the TDCR method. I eliminated the initial inaccuracy of the calculation by including the scintillation effects of the Compton photons scattering. Determination of the activity using the counting code was difficult as one value of the specific parameter of the TDCR method in the experimental range corresponded to three different values of counting efficiency. I suggested using the counting efficiencies of triple coincidence to a logical sum of the double coincidence in the third power ratio as a more convenient experimental parameter of the TDCR method for the ^{139}Ce calculation. The ^{139}Ce counting efficiency could be presented as a univocal function of this parameter in the experimental range. A set of ^{139}Ce sources in a liquid scintillator was prepared and measured using the TDCR method. A set of counting points, which I calculated, matched very well the ^{139}Ce counting efficiencies. Using the obtained coincidence counting rate, I determined the radioactive concentration of the ^{139}Ce solution with a standard uncertainty of ± 0.5 %. The same ^{139}Ce solution was standardized using the TDCR method also in the LPRI. A very high consistency of the measurement results in both laboratories was obtained. The determined value of the radioactive concentration of the ^{139}Ce solution was confirmed by other measurement methods.

Conclusions related to the method of calculation of the ^{139}Ce counting efficiency in the triple counter with a liquid scintillator were significant for the subsequent development of the TDCR method.

2. **Broda R.**, Cassette P., Małetka K., Pochwalski K. A simple computing programme for the application of the TDCR method to standardization of pure-beta emitters, *Appl. Radiat. Isot.*, 52, (2000) 673-678.

Growing interest in the TDCR method and construction of triple counters in various metrological laboratories all over the world required availability of the basic computing code for the calculation of the counting efficiency of pure β-emitters in the triple counter. I prepared the TDCRB-1 code for publication that utilized the French subprogram for the calculation the shape of beta spectra, which is a useful tool for the user. The TDCRB-1 code worked in an interactive mode and enabled us to determine the radioactive concentration of a solution with a measurement uncertainty or the counting efficiency of a given radionuclide in a selected range of input parameters. The modular configuration of the code allowed as and where necessary easy modification of the subprograms and matching the input data format to the requirements of a given triple counter. I gathered nuclear and atomic data for ten different β-emitters and several kinds of liquid scintillators necessary for the calculations in a separate file. I introduced a graphical representation of the computing results to enable their quick assessment by the user. I described the algorithm of the TDCRB-1 code and the input/output data format. I prepared the main part of the description of the calculation model and assumptions made with respect to working conditions of the TDCR detector. One source of the ^{63}Ni solution was measured using different counting efficiencies. The radioactive concentration of the ^{63}Ni solution using the TDCRB-1 code was determined. The User Manual was prepared using all the sets of input and output data including data format descriptions.

3. Cassette P., **Broda R.**, Hainos D., Terlikowska T. An analysis of detection-efficiency variation techniques for the implementation of the TDCR method in liquid scintillation counting, *Appl. Radiat. Isot.*, 52, (2000) 643-648.

It was observed that the main source of uncertainty regarding the activity determined by the TDCR method especially in the case of low-energy radionuclides, is the Birks function [Broda et al., 1988] applied in the model of the LS-counter detection efficiency calculation with a selected ionization quenching parameter, i.e. *kB*. The function describes the scintillation light emission as a non-linear function of electron energy due to the ionization quenching. Variations in the triple counter detection efficiency are an important element of the measurement procedure allowing the assessment of how correct the computing model and the *kB* parameter value is. The value of the radionuclide activity determined in various counting conditions should fulfil the criterion of counting correctness . For correct application of the calculation model in the TDCR method, the influence of different ways of counting efficiency variations on the triple counter measurement results had to be verified.

A change in the detection efficiency can be achieved by increasing the chemical quenching in the scintillator e.g. by adding nitromethane, or using coaxial optical filters around the liquid scintillator vial or PMTs defocusing, i.e. reducing their efficiency by decreasing the focusing electrode potential. In the first two cases, the number of registered scintillation photons is reduced. In the LWR laboratory practice we applied the PMTs defocusing as a most convenient way of changing the counting efficiency of the triple counter [Broda and Pochwalski, 1992]. Measurements of the ^{3}H, ^{14}C and ^{63}Ni sources activity using the TDCR method while changing the counting efficiency in the three above-given ways were carried out in the LPRI, France. My calculations which were based on the TDCRB-1 code produced identical results showing that all of the applied quenching methods were equivalent while the *kB* parameter was independent of the chemical quenching degree and the optical properties of the detector. The counting efficiency model did not have to be modified for different quenching methods.

Based on the results of the calculations, I showed that the applied calculation model failed for low detection efficiency when the number of the emitted scintillation photons was low and it was correct only when an expected mean number of photoelectrons per one PMT exceeded 1. I also showed that the calculated values of low-energy radionuclides ^{3}H and ^{63}Ni activity were strongly dependent on the *kB* parameter value adopted whereas the influence of this parameter on the calculated value of activity of high-energy emitter ^{14}C was negligible. It was the result of a strong nonlinearity of the Birks function for electron energies below about 20 keV. For all sources of the three radionuclides standardized in the LPRI, I found the same value of the ionization quenching parameter, i.e. $kB \approx 0.011$ cm MeV^{-1} using the criterion of the counting correctness and the Poisson statistics for photons collected by PMTs. One could suppose that the identified *kB* value was specific to the Ultima Gold liquid scintillator used.

4. **Broda R.**, Małetka K., Terlikowska T., Cassette P. Study of the influence of the LS-cocktail composition for the standardisation of radionuclides using the TDCR model, *Appl. Radiat. Isot.*, 56, (2002) 285-289.

Previous works showed that correct determination of the activity of the radionuclide measured by the TDCR method depends on the use of a proper value of the ionization quenching parameter *kB* in the calculations. The *kB* parameter value was not known *a priori*. As the Poissonian model was used to describe the statistics of scintillation photons, it was convenient to take the *kB* parameter as a variable to match the calculated counting efficiency to a set of counting points in each source. However, this method of the source activity and the *kB* parameter determination was not physically justifiable.

The results of the previous work (i.e. [3]) did not provide an answer to the question whether the *kB* parameter value varies depending on the type of the liquid scintillators and their composition. To verify this I planned new measurements. A set of ^{3}H and ^{63}Ni sources using standard solutions in four different commercial liquid scintillators as well as in the home-made toluene and seven xylene scintillators with varying levels of xylene to 2-ethoxyethanol solvents ratios was prepared. The activity of all sources was determined by means of the Wallac 1411 scintillation spectrometer noting at the same time that all of the above-mentioned scintillation cocktails differed in the chemical quenching levels.

I developed an improved TDCRB-02 code for calculating the counting efficiency and determining the radionuclide activity using the TDCR method in the triple counter. I used the Downhill Simplex multidimensional minimization method to solve the system of three equations with experimental data. Taking the ^{3}H activity measurement as an example, I justified the necessity to take into account three different PMT optical efficiencies in calculations. The TDCRB-02 code was thrown open to users at the web site of the Liquid Scintillators Working Group of the International Committee for Radionuclide Metrology (ICRM). Having performed the measurements of ^{3}H and ^{63}Ni standard sources by means of the TDCR method with the triple counter, I managed to determine the counting efficiency in each case. I calculated the *kB* parameter value taking into consideration the chemical composition of each of the scintillation cocktails used. In the case of commercial scintillators, whose composition constituted a manufacturer secret, I used the published data concerning the mass fraction of atoms of the particular components of the scintillation cocktail. I showed that the ionization quenching parameter *kB* for all scintillation cocktails used in measurements of the ^{3}H and ^{63}Ni solutions had a constant value of about 0.011 cm MeV^{-1} and fell within the uncertainty limits ± 0.001 cm MeV^{-1} without being dependent on the chemical composition of these cocktails. These findings were confirmed by the measurements performed in the Laboratoire National Henri Becquerel (LNHB, former: LPRI).

5. Broda R., Jęczmieniowski A. Statistics of the LS-detector in the case of low counting efficiency. *Appl. Radiat.Isot.*, 60, (2004) 453-458.

The results published by Cassette et al. [3] showed that the calculated LS-counter detection efficiencies for high-quenched ^{3}H and ^{63}Ni sources were inconsistent with the experimental values. The description of the detection process by the Poisson distribution was incorrect when an expected mean number of photoelectrons per one PMT was smaller than 1. I decided to investigate the photoelectrons statistics for low-energy ^{3}H and ^{55}Fe emitters using the TDCR method. I measured the radionuclides by PMTs defocusing and using optical filters for variation the triple counter detection efficiency. I developed counting codes to determine the detection efficiencies with various statistical distributions for photoelectrons. The computed results confirmed that the calculation model could not be applied when the detection efficiency was low and the expected mean number of photoelectrons per one PMT was lower than 1. I showed also that in the calculation of the ^{3}H and ^{55}Fe activities, even with higher detection efficiencies the criterion of the counting correctness was not satisfied. Different calculated activities were obtained for various LS-counter detection efficiencies when the Poisson distribution for photoelectrons in the calculation model was used. The calculated mean ^{3}H and ^{55}Fe activity values were about 2 % and 5 % lower respectively. I obtained the correct results of the above radionuclides standardization using the binomial negative distribution (Polya distribution).

I measured the ^{55}Fe, an almost monoenergetic EC-nuclide, and compared it with the results of the LED simulated the monoenergetic scintillation source. The LED was inserted into the triple counter and excited by a pulse generator synchronized by a signal from other LS-counter. In this way the pulse generator became a random pulse generator. I also measured the LED frequency using the TDCR method and PMTs defocusing, using optical filters or reducing the LED pulse amplitude. I developed the code for the LED frequency calculation. The thus obtained mean LED frequency was correct when the Poisson distribution for photoelectrons was used. The same mean LED frequency value for various PMTs defocusing and detection efficiencies was obtained when the expected number of photoelectrons per one PMT exceeded 1. Both the ^{55}Fe and LED could be regarded as monoenergetic sources, but various statistical distributions for photoelectrons had to be used. I formulated the conclusion that the liquid scintillator which was not present in the LED measurements distorted the ^{55}Fe calculation results when the Poisson distribution was used. The Polya distribution should be used in the ^{55}Fe activity calculation.

6. Broda R. Some remarks on photons statistics in the LS-counter. *Appl. Radiat. Isot.*, 66, (2008) 1062-1066.

The conclusion that the correct low-energy ^{3}H and ^{55}Fe radionuclides standardization needs the Polya instead the Poisson distribution for PMT-collected photons was not physically justifiable [5]. I tried to interpret this fact based on the analysis of the processes leading to fluorescence and the light emission from LS-vials data (Cassette P., Vatin R. *Experimental evaluation of TDCR models for 3 PM liquid scintillation counter. Nucl. Instr. Meth. Phys. Res.* A 312, (1992) 95-99).

I found the probability values of the successive processes leading to fluorescence in the liquid scintillator in the literature and presented them in a simplified graphical form. I calculated that in a single decay of ^{3}H or ^{55}Fe nucleus, about 20 scintillation photons are emitted on an average. The emitted photons could be described by the binomial or the Poisson distribution approximately. Cassette and Vatin (1992) showed, however, that the global statistics of the PMT-collected photons showed larger variances than the two above-mentioned distributions. The authors explained that the underlying reasons were the internal light reflections at the interface between the LS-vial wall and the air. They obtained the LS-vial light emission probability as a function of the light emission position in the LS-cocktail using a Monte Carlo simulation. These data allowed me to calculate the approximate shape of the PMT-collected photons statistical distribution. I calculated the binomial distributions for photons emitted from different layers of the liquid scintillator and summarised them taking into account the volume of each layer. I showed that the shape of global distribution could be satisfactorily approximated by the Polya distribution for equal expected number of photons. Using the Polya distribution definition I also demonstrated that its variance was larger than the binomial or the Poisson distribution. The above results justified the use of the Polya distribution in the LS-counter detection efficiency calculation model for low-energy radionuclides.

7. **Broda R.** A review of the triple-to-double coincidence ratio (TDCR) method for standardizing radionuclides. *Appl. Radiat.Isot.*, 58, (2003) 585-594.

After twenty years of development of the absolute TDCR method, I reviewed the published results related to the application of this method in radionuclide standardizing. The review was to present the current state of development of the TDCR method and interest it aroused in a number of laboratories across the world. I described how the method was improved. I pointed out that adopting the triple counter version proposed by K. Pochwalski and calculating the counting efficiency using the model based on a statistical description of the phenomena in the LS-detector were the most important factors in this process. I summarized the theoretical grounds of the TDCR method and presented the scheme for the calculation of the counting probability in the LS-counter. The initial development of the TDCR method was connected with the Polish – French scientific cooperation and progress in the coincidence modules used in the triple counter construction. Simplified block-diagrams of the counters were presented. I collected the details of the triple counters constructed in twelve different laboratories across the world emphasizing the structural differences between them.

To demonstrate how different the counting efficiencies of various radionuclides in the triple counter can be, I performed the calculation for some β-emitters and EC-nuclides. I presented the calculated efficiencies as a function of a specific parameter of the TDCR method in a graphical form. I explained that the apparent differences in the counting efficiency curves depended on the differences in the shape of the scintillation spectra of different radionuclides. In the case of the EC-nuclides, the more separate groups of energy can be distinguished in the scintillation spectrum as a result of X- and γ-rays interaction with the scintillator and the atomic rearrangements after electron capture, the more complicated the curves become. It could be observed for example in ^{139}Ce [1]. I justified the conclusions with the calculation of the counting efficiency of a simplified spectrum consisting of two selected energies. I provided examples of activity measurement results of some radionuclides by means of the TDCR method indicating that the criterion of the counting correctness was satisfied.

8. **Broda R.**, Cassette P., Kossert K. Radionuclide metrology using the liquid scintillation counting. *Metrologia*, 44, (2007) S36-S52.

To show the significance of the TDCR method and the detection efficiency calculation model in a wider context of the radionuclide metrology using the LSC technique, I participated in a joint effort to prepare the chapter devoted to the LSC technique in a special issue of the *Metrologia* bimonthly journal. This issue was compiled upon the initiative of the Consultative Committee for Ionizing Radiation (CCRI) of BIPM by the most competent and experienced metrologists from various countries. The issue presented the "state of art" developments and progress in ionization radiation metrology. It was conceived as a resource for new metrologists joining the field and a guide for radioactivity users explaining how traceability to the international measurement system may be achieved.

In the chapter devoted to the use of the LSC technique, the importance of measuring practice was emphasised. The processes responsible for conversion of the ionizing radiation energy into fluorescent light emitted by the excited molecules of scintillator were presented as well as the composition of the scintillation cocktail with an aqueous radioactive solution required for an effective transfer of the energy from the solvent to the scintillator molecules. In the discussion of the quenching processes in the scintillator, which reduce the number of emitted fluorescent photons, the fluorescence efficiency calculation using the Birks function was presented. Also, the calculation of the detection efficiency in the LS-counters was discussed in detail [Broda et al., 1988]. The above-mentioned calculations require prior determining of an exact electron energy spectrum that appears in the scintillator as a result of the radioactive decay. The calculation of the electron spectrum after the beta decay, the electron-capture, interaction of X or γ photons with the scintillator were demonstrated as well as the way of calculating the electrons spectra of radionuclides with a complex decay scheme.

The CIEMAT/NIST efficiency tracing method using tritium as a tracer and the influence of selection of the tracer on measurement accuracy were discussed in reference to practical utilization of the LSC technique for the measurements of radionuclides activity with the double counter. Furthermore, the TDCR method and the way of determining the radionuclide activity based on the counting efficiency calculation were presented [Broda and Pochwalski, 1992]. It was emphasised that without the above calculations, the linear extrapolation of counting points may lead to incorrect results especially in the case of the EC-nuclides. The conclusions related to the value of the *kB* ionization quenching parameter for the liquid scintillators used were put together. Various radionuclides standardized through the application of both of the above methods were discussed. In the overview of practical aspects of the LSC technique, the features required in the measurement vials and the liquid scintillators for an effective employment the both measurement methods were identified as well as the kinds of counters used, counter testing methods and the ways to change the counting efficiency to allow assessment of the efficiency calculation accuracy. It was emphasized that radionuclidic impurities of the measured solutions can distort the calculation results. The amount of the γ-impurities in a given solution can be determined using a standard method of gamma spectrometry. The β-impurities are often difficult to detect.

My contribution to the joint work consisted in particular in describing the calculation model of the detection efficiency in the LS-counters and demonstrating its employment in the TDCR method. I presented derivations of formulae for β-emitters and EC-nuclides counting efficiency in the LS-counters with various number of PMTs. Furthermore, I provided probabilities of registration of various kinds of pulses based on the assumption that PMTs are either identical or non-identical. I also demonstrated that correct determination the activity of low-energy ^{3}H and ^{55}Fe radionuclides by means of the TDCR method requires binomial negative distribution (Polya distribution) for scintillation photons collected by PMTs. Alternatively, the use of the Poisson distribution provides the ^{3}H and ^{55}Fe activity values which are about 2 % and 5 % lower respectively. As it turned out, the reason was the internal light reflections on the walls of glass vials containing the scintillation cocktail. I demonstrated the detection efficiencies of various radionuclides in the function of the specific experimental parameter of the TDCR method and how to calculate them in the case of non-identical PMTs in the triple counter.

Recapitulation

The research results described in the above-quoted publications shows the way of the LS-counter detection efficiency calculation model application for various radionuclides activity determination using the TDCR method. I presented conclusions concerning the LS-counter detection efficiency variations occurring during the radionuclide activity measurements. I determined the ionization quenching parameter value and the statistical distribution for the photoelectrons applied in the calculation model. I also showed that the applied calculation model failed for low detection efficiency in the triple counter.

Changing the counter detection efficiency by one of the three methods below, i.e. chemical quenching, optical filters application or, PMTs defocalization being the most convenient method usually applied in activity measurement of any radionuclide by means of the TDCR method. The goal of the efficiency change is to register the counting points enabling the estimation of the correctness of the applied calculation model where the ionization quenching parameter is one of variables. Its value, which is not known *a priori*, strongly influences the determined activity of the low-energy β-emitters. The test measurements of a set of ^{3}H, ^{14}C and ^{63}Ni sources showed, that all of the above methods of the detection efficiency change are equivalent and the ionization quenching parameter has a fixed value of about 0.011 cm MeV^{-1}. It was also found that the value is stable regardless of the chemical composition of the scintillation cocktail, which was confirmed by the measurements carried out in France. The above observations were of considerable importance for the practice of the TDCR method application. Also essential were the conclusions concerning the applied calculation model. While performing the low-energy β-emitters measurements I noticed that the developed model has a limitation. It cannot be applied if the counter efficiency is low and the expected number of emitted photoelectrons per one PMT is lower than one. The obtained calculation results do not match the experimental points then. The model allowed me to calculate the counting efficiency of various radionuclides such as the EC-nuclides as a function of specific parameter of the TDCR method. I demonstrated that the functions depend on the shape of the scintillation spectra of particular radionuclides. I justified the conclusions with the calculations concerning a virtual radionuclide with a very simplified scintillation spectrum.

Test measurements of the low-energy emitters ^{3}H and ^{55}Fe showed, that in the calculation of their activity the binomial negative distribution (Polya distribution) instead of the Poisson one for the PMT-collected photons should be applied. The reason was the internal light reflection in the walls of LS-vials. In the case of test measurements with the LED diode simulating the scintillation source and fixed axially in the vial without a scintillator, the light reflections did not occur and the Poisson distribution in the calculated model was applied.

The joint LWR and LPRI works showed, that the developed calculation model of the LS-counter detection efficiency can be applied to the γ-emitting radionuclides activity measurements by means of the TDCR method too. I analyzed the ^{139}Ce decay scheme which was selected to test the model and determined the spectrum of electrons emitted following the ^{139}La atom shells rearrangement including the scintillation effects of the Compton photon scattering. I obtained the ^{139}Ce counting efficiency calculation results which were in agreement with the experimental efficiency. The LWR and LPRI results of measurements of the ^{139}Ce solution radioactive concentration were consistent and the measurement standard uncertainty was estimated at ± 0.5 %.

The TDCR method implementation required the triple counter and the computation code based on the developed calculation model. I published the TDCRB-1 code for activity calculation of the pure β-emitters, and then the TDCRB-02, which is an improved version of the code accessible at the ICRM Working Group for Liquid Scintillators website. I published a review of articles referring to the TDCR method and to the history of its improvement to provide the reader with the basic data and the information on the way of implementation in a dozen or so laboratories. I also participated as the co-author in the preparation of a comprehensive and detailed publication presenting the TDCR method in the context of the LSC technique application with the emphasis on the measurement practice and the application of the LS-counter detection efficiency calculation model . The developed calculation model for the TDCR method and the formulated conclusions were used in the LWR laboratory practice. . The TDCR method was successfully applied in the international comparisons of β-emitter ^{63}Ni [Razdolescu et al., 2006] and radioactive concentration of the ^{99}Tc solution calculation [Zimmerman et al., 2010] as well as in the activity of the ^{85}Sr being the EC-nuclide with a delayed isomeric transition [Broda et al., 2012]. The development of the physical basis of the TDCR method and creation of the calculation model as well as dozens of publications concerning the method and its advantages as an absolute method contributed to its introduction into the international reference system which was extended to include the β-emitters measurements (ESIR) at BIPM, France.

5. OTHER R&D ACHIEVEMENTS

5.1 Other research and development activity

In addition to the issues related to the above-indicated scientific achievement, I was also involved in other research and development problems, which I describe below. Initially, my activity concerned the relative methods of the radionuclides activity measurements by means of the ionizing chambers applied to the control of sources and radioactive solutions produced at RC POLATOM in Świerk. I analyzed the ionization chambers parameters and determined the counting efficiency of particular radionuclides. I developed the method of the ^{99m}Tc activity determination in ionization chamber based on the ^{99}Mo activity measurements in the $^{99}Mo+^{99m}Tc$ solution [Broda, 1980]. I participated in development of the sealed radioactive sources activity measurement methods in ionization chambers taking into account the radiation self-absorption in the source. The result of the work was identification of a suitable geometry of source measurement and ionization chamber calibration.

My activity was also related to the sealed sources absorbed dose rate measurement by means of an extrapolation chamber. I was the co-author of the "HMD, Sealed radioactive sources of radiation. Methods of measuring the absorbed dose rate of beta-radiation" standard (in Russian), Moscow (1980) as well as the co-author of the trade standard entitled: "Sealed radioactive sources: marking and attestation" (in Polish), BN-77, 3421-01 (1977). Within the framework of contacts with the nuclear medicine units, which are the main users of radioactive solutions produced in RC POLATOM, I developed and patented a system of convenient lead covers for syringes containing a radioactive solution [R. Broda, A. Grabowski, Utility Pattern No. 55774: "Lead cover for syringes containing a radioactive solution", Patent Office, Republic of Poland, Warsaw, 1988].

Another field of my activity was application of the liquid scintillation technique in the process of determination of the radioactive concentration of various solutions. I participated in collective works on the afterpulses in the LS-counter that distorted the results of activity measurement [Broda and Radoszewski, 1979]. The cause of the above-mentioned pulses were the true pulses occurring after the radioactive decay in the source. We found that the afterpulses are one-electron mainly and we determined their amplitude spectrum and time distribution after the true pulse. Application of a long dead time of the order of 40 μs and of the extendible dead time in the improved LS-counters enabled us to avoid the detection of those pulses.

Other works were related to the investigation of the reasons underlying the observed differences in the international comparisons of the radioactive concentration of the ^{55}Fe, ^{63}Ni [Cassette et al., 1998] and ^{3}H [Makepeace et al., 1998] solutions results. A correlation between obtained results and the scintillation cocktails used as well as the dependence on the TDCR or the applied CIEMAT/NIST measurement method was observed. In collaboration with the LPRI laboratory, I participated in investigation of the stability of the ^{63}Ni sources prepared in the Ultima Gold® scintillation cocktail [Terlikowska et al., 1998]. My contribution in the works consisted in performing the measurements of the radioactive concentration of the ^{63}Ni solutions by means of the TDCR method. The way of the sources preparation was changed. Different results of measurements of the activity of the sources prepared in various lots of the Ultima Gold® scintillator were obtained. Distinctly different results were obtained depending on the type of the scintillator used, i.e. Ultima Gold ® or Ultima Gold ® AB . It was found that the sources containing more than 1.5 % aqueous phase were characterized by the better stability.

One of the last problems I dealt with was the ^{85}Sr measurement by means of the TDCR and CIEMAT/NIST method [Broda et al., 2012]. ^{85}Sr decays mainly by electron capture to the excited level of ^{85}Rb having the half-life of about 1 μs and following by the isomeric transition to the ground level. I observed a clear divergence of the measurement results obtained by both of the above-mentioned methods. I found an explanation of this fact. I ascertained that the reason was lack of a part of scintillation pulses registration, i.e. the loss of pulses, as the dead time fixed in the LS-counter was much longer than the half-life of the excited level. After a suitable correction in the calculation model and using both methods I obtained consistent results within the acceptable measurement uncertainty.

My scientific activity also involved preparation of 16 reviews of papers submitted for publication in scientific journals *Applied Radiation and Isotopes* and *NUKLEONIKA*.

5.2 Publications

A) Indexed publications

Number of indexed publications: **22**

A1) Indexed publications before the doctor's degree

Number of indexed publications before doctoral thesis: **4**

Pochwalski K., Broda R., Radoszewski T. Standardization of pure beta emitters by liquid - scintillation counting. *Appl. Radiat. Isot.*, 39, (1988) 165-172.

Broda R., Pochwalski K., Radoszewski T. Calculation of liquid - scintillation detector efficiency. *Appl. Radiat. Isot.,* 39, (1988) 159-164.

Broda R., Pochwalski K. The enhanced triple-to-double coincidence ratio (ETDCR) method for standardization of radionuclides by liquid-scintillation counting. *Nucl. Instr. Meth. Phys. Res.*, A312, (1992) 85-89.

Broda R., Pochwalski K. The ETDCR Method of standardizing ^{55}Fe and ^{54}Mn. liquid-scintillation spectrometry 1992. ed.: Noakes J.E., Schönhofer F., Polach H.A., *RADIOCARBON*, USA. (1993) 255-260.

A2) Indexed publications after the doctor's degree

Number of indexed publications after doctoral thesis: **18**

Cassette P., Altzitzoglou T., Broda R., Collé R., Dryák P., De Felice P., Günther E., Los Arcos J.M., Ratel G., Simpson B., Verrezen F. Comparison of activity measurement of ^{63}Ni and ^{55}Fe within the scope of the EUROMET 297 Project. *Appl. Radiat. Isot.,* 49, (1998) 1403-1410.

Makepeace J., Altzitzoglou T., Cassette P., Dryák P., Günther E., Verrezen F., Broda R., Simpson B., Unterweger M. International comparison of measurements of the specific activity of tritiated water. *Appl. Radiat. Isot.,* 49, (1998) 1411-1416.

Terlikowska T., Cassette P., Péron M., Hainos D., Tartes L., Broda R., Kempisty T. Study of stability of ^{63}Ni sources in Ultima Gold liquid scintillation cocktail. *Appl. Radiat. Isot.,* 49, No. (1998) 1041-1047.

Broda R., Péron M., Cassette P., Terlikowska T., Hainos D. Standardization of ^{139}Ce by liquid scintillation counting using the triple to double coincidence ratio method. *Appl. Radiat. Isot.,* 49, (1998) 1035-1040.

Cassette P., Broda R., Hainos D., Terlikowska T. Analysis of detection-efficiency variation techniques for the implementation of the TDCR method in liquid scintillation counting. *Appl. Radiat. Isot.*, 52, (2000) 643-648.

Broda R., Cassette P., Małetka K., Pochwalski K. A simple computing program for application of the TDCR method to standardization of pure-beta emitters. *Appl. Radiat. Isot.*, 52, (2000) 673-678.

Broda R., Małetka K., Terlikowska T., Cassette P. Study of the influence of the LS-cocktail composition for the standardisation of radionuclides using the TDCR model. *Appl. Radiat. Isot.*, 56, (2002) 285-289.

Chyliński A., Terlikowska – Droździel T., Radoszewski T., Broda R. Multi-method of standardisation of radionuclides with triangular scheme of disintegration. *Appl. Radiat. Isot.*, 56, (2002) 281-284.

Chyliński A., Broda R., Radoszewski T. The national standard unit of radionuclide activity and the related standards in Poland. *NUKLEONIKA*, 48 (1), (2003) 51-55.

Broda R. A review of the triple-to-double coincidence ratio (TDCR) method for standardizing radionuclides. *Appl. Radiat.Isot.*, 58, (2003) 585-594.

Broda R., Jęczmieniowski A. Statistics of the LS-detector in the case of low counting efficiency. *Appl. Radiat.Isot.*, 60, (2004) 453-458.

Razdolescu A.C., Broda R., Cassette P., Simpson B.R.S., Van Wyngaardt W.M. The IFIN-HH triple coincidence liquid scintillator counter. *Appl. Radiat.Isot.*, 64, (2006) 1510-1514.

Broda R., Cassette P., Kossert K. Radionuclide metrology using liquid scintillation counting. *Metrologia*, 44, (2007) S36-S52.

Broda R. Some remarks on photons statistics in the LS-counter. *Appl. Radiat. Isot.*, 66, (2008) 1062-1066.

Zimmerman B.E., Altzitzoglou T., Rodrigues D., Broda R., Cassette P., Mo L., Ratel G., Simpson B., Van Wyngaardt W., Wätjen C. A comparison of triple-to-double coincidence ratio (TDCR) efficiency calculations and uncertainty assessments for ^{99}Tc. *Appl. Radiat. Isot.*, 68, (2010) 1278-1281.

Broda R., Dziel T., Muklanowicz A., Listkowska L., Pieńkowski Ł., Patocka A., Kołakowska E. A comparison of ^{99}Tc^{m} and ^{131}I in Polish hospitals, 2007. *Appl. Radiat. Isot.*, 68, (2010) 1278-1281.

Broda R., Dziel T., Muklanowicz A. Standardization of a ^{85}Sr solution by three methods. *Appl. Radiat. Isot.*, 70, (2012) 2222-2226.

Dziel T., Broda R., Ziemek T., Muklanowicz A., Listkowska A. Standardization of Sm-153 solution by absolute methods. *Appl. Radiat. Isot.*, 87, (2014) 19-23.

B) Other publications (reports, non-indexed publications, proceedings)

Number of other publications: **18**

B1) Other publications before the doctor's degree

Number of other publications before doctor's degree: **9**

Broda R., Radoszewski T. Study of afterpulses in a liquid scintillation counter (in Russian). Proceedings on Symposium on the "Methods of manufacturing and measuring standard sources and solutions", Marianske Lazne 1979, Czechoslovakian Atomic Energy Commission, Prague, Czechoslovakia (1979).

Broda R. The method of activity determination of ^{99m}Tc in ionization chamber, INR 1870/OPiDI/I/A, Warsaw, Poland (1980).

Pochwalski K., Broda R., Radoszewski T., Żelazny P. Standardization of low-level beta-emitter solutions by using the liquid scintillation triple-to-double coincidence ratio (TDCR) method. "Methods of low-level counting and spectrometry", IAEA-SM-252/62, Vienna, Austria, (1981) 487-496.

Broda R., Pochwalski K. Calculation of the liquid-scintillation detector counting efficiencies in comparison with the experimental data. INR 1942/OPiDI/E/A, Warsaw, Poland (1982).

Broda R., Radoszewski T. Scintillation detector with anticoincidence shield for determination of the radioactive concentration of standard solutions. The second International Conference on "Low activities", High Tatras 1980, *Physics and Applications*, vol. 8, VEDA, Bratislava, Czechoslovakia, (1982) 156-167.

Broda R., Chyliński A., Terlikowska-Droździel T., Kempisty T., Pochwalski K., Radoszewski T. Developments in the scope of metrology of the standard radioactive sources (in Russian). Proceedings on the second COMECOM Symposium on the "Methods of manufacturing and measuring standard sources and solutions". Hungarian Atomic Energy Commission, Copak, Hungary, (1984) 189-223.

Broda R. Theoretical determination of the measurement efficiency of ^{55}Fe in liquid scintillator in comparison with the experimental data (in Russian), Proceedings on the second COMECOM Symposium on the "Methods of manufacturing and measuring standard sources and solutions". Hungarian Atomic Energy Commission, Copak, Hungary, (1984) 271-277.

Pochwalski K., Broda R., Radoszewski T. Absolute measurement of pure beta-emitters using the liquid scintillator counter, (in Polish), INT Report 198/1, Symposium on the "Development and application of isotopic methods in technology", AGH, Kraków, (1986).

Broda R. WYDA16 and ECFE11 codes for radioactive concentration calculation of solutions measured by the triple-to-double coincidence ratio (TDCR) method. Report IAE 2081/OPiDI/I/B, Świerk, Poland (1989).

B2) Other publications after the doctor's degree

Number of other publications after doctor's degree: **9**

Chyliński A., Broda R., Radoszewski T. The national standard unit of radionuclide activity, (in Polish). *Postępy Techniki Jądrowej*, Vol. 44, Z.2, (2001) 24-31.

Broda R. The National standard of radionuclide activity unit. (in Polish), *Pomiary Automatyka Robotyka*, No.7-8, (2003) 51-55.

Ratel G., Michote C., Broda R., Listkowska A. Activity measurements of the radionuclide ^{60}Co for the RC, Poland in the ongoing comparison BIPM.RI(II)-K1.Co-60. BIPM, Report-Co-60 (3), 2003/09/09.

Zimmerman B.E., Colle R., Cessna J.T., Broda R., Cassette P. Application of the triple-to- double coincidence ratio method at the National Institute of Standards and Technology for absolute standardization of radionuclides by liquid scintillation counting. *Radioanalytical Methods In Interdisciplinary Research*. ACS Symposium Series 868, American Chemical Society, Washington, DC, (2004) 76-87.

Broda R., Małetka K., Muklanowicz A., Listkowska A. Metrological laboratory in RC POLATOM. Proceedings on the national conference on "Nuclear technique in industry, medicine, agriculture and environmental protection", (in Polish), Stanislaw Staszic University of Science and Technology (AGH), Kraków, (2005) 549-554.

Broda R., Chyliński A., Radoszewski T., Małetka K., Terlikowska-Droździel T. The national standard of the radionuclides activity unit in Poland. Proc. Internat. "Applications of high precision atomic & nuclear methods" Conference, Ed.: Acad. Romane, Rumunia, (2005) 63-68.

Dziel T., Broda R. The national standard of the radionuclides activity unit, (in Polish). *Metrologia.* Information Bulletin of the Central Office of Measures. 12, (2009).

Broda R., Pochwalski K. The origin and evolution of Laboratory of Radionuclide Metrology in Świerk, (in Polish). *Postępy Techniki Jądrowej, 54,* (2011) *14-21.*

Gajda J., Broda R., Jachowicz R., Mroczka J. et al. The status and perspectives of development of the metrology as a scientific inter-discipline (on a global scale). The Committee for Metrology and Scientific Instrumentation of the Polish Academy of Science in the 2010 – 2014 tenure activity guide-book. [in Polish]. Zielona Góra (2014) 41-72.

5.3 Quotations

Total *Impact factor* of publications by *Journal Citation Reports* (JRC), in the year of publication: **17.8**

Quotations according to the Web of Science base:
- Total number of quotations: **282**
- Total number of quotations without self-quotation: **227**

Hirsch Index according to the Web of Science base: **9**

5.4 Participation in conferences

A) Lectures presented by R. Broda at international conferences:

1979 COMECOM Symposium on the "Methods of manufacturing and measuring standard sources and solutions", Mariańskie Łaźnie, Czechoslovakia – Broda R., Radoszewski T. Study on afterpulses in a liquid scintillation counter (in Russian). – published in conference proceedings.

1980 Second International Conference on "Low activities", Strbskie Pleso, Czechoslovakia – Broda R., Radoszewski T. Scintillation detector with anticoincidence shield for determination of the radioactive concentration of standard solutions. – published in conference proceedings.

1984 Second COMECOM Symposium on the "Methods of manufacturing and measuring standard sources and solutions", Copak, Hungary – Broda R. Theoretical determination of the measurement efficiency of ^{55}Fe in liquid scintillator in comparison with experimental data (in Russian). – published in conference proceedings.

1985 5th ICRM Conference and Working Group Meeting, Grenoble, France – Broda R. Some remarks on statistical distribution of photons and photoelectrons in liquid scintillation detectors.

1987 6th ICRM Conference, Rome, Italy – Broda R., Pochwalski K., Radoszewski T. Standardization of the ^{109}Cd-^{109m}Ag solution by a triple-to-double coincidence ratio method.

1991 8th ICRM Conference on the "Radionuclide metrology and its application", Madrid, Spain - Broda R., Pochwalski K. The enhanced triple-to-double coincidence ratio (ETDCR) method for standardization of radionuclides by liquid-scintillation counting. - published in *Nucl. Insr. Meth. Phys. Res.*

1992 International Conference on the "Advances in liquid-scintillation spectrometry", The Austrian Society for Liquid-Scintillation Spectrometry, Vienna, Austria – Broda R., Pochwalski K. The ETDCR method of standardizing ^{55}Fe and ^{54}Mn. - published in

RADIOCARBON.

1997 11th ICRM Conference on the "Radionuclide metrology and its application", Gaithersburg, Md. USA - Broda R., Péron M., Cassette P., Terlikowska T., Hainos D. Standardization of ^{139}Ce by liquid scintillation counting using the triple to double coincidence ratio method. - published in *Appl. Radiat. Isot.*

1999 Conference of the Korean Physical Society, Pusan, Republic of Korea – Broda R. Application of liquid scintillation counting (LSC) for radionuclide activity determination.

2001 13th ICRM Conference on the "Radionuclide metrology and its application", Braunschweig, Germany – Broda R., Małetka K., Terlikowska T., Cassette P. Study of the influence of the LS-cocktail composition for the standardisation of radionuclides using the TDCR model. - published in *Appl. Radiat. Isot.*

2002 International Conference of the European Commission Centre of Excellence Interdisciplinary Research and Applications Based on Nuclear and Atomic Physic (IDRANAP) on the "Applications of high precision atomic & nuclear methods", Neptun, Romania – Broda R., Chyliński A., Radoszewski T., Małetka K., Terlikowska-Droździel T. The national standard of the radionuclides activity unit in Poland. – published in conference proceedings.

2003 14th ICRM Conference on the "Radionuclide metrology and its applications", ICRM 2003, Dublin, Ireland – Broda R., Jęczmieniowski A. Statistics of the LS-detector in the case of low counting efficiency. - published in *Appl. Radiat. Isot.*;

2005 12th Meeting of EUROMET QS-Forum, Bucharest, Romania – Broda R. Presentation of the Quality System.

2005 15th ICRM Conference on the "Radionuclide metrology and its applications", ICRM 2005, Oxford, United Kingdom – Broda R., Dziel T. Technical Comparison of ^{63}Ni using the TDCR method (A preliminary analysis).

2009 4th Meeting of EURAMET TC-Quality, Thessaloniki, Greece – Broda R. Presentation of the QMS re-evaluation.

2011 18th International Committee for Radionuclide Metrology (ICRM) Conference on the "Radionuclide metrology and its applications", ICRM 2011, Tsukuba, Japan – Broda R., Dziel T., Muklanowicz A. Standardization of a ^{85}Sm solution by three methods; - published in *Appl. Radiat. Isot.*; - Chairman of the session.

B) Lectures presented by R. Broda at national conferences:

1980 First National Liquid-Scintillation Technique Workshop, Kołobrzeg.

1985 A National Symposium AGH on the "Development and implementation of isotope methods in engineering and technology", Zakopane – Pochwalski K., Broda R., Radoszewski T. An absolute measurement of pure beta-emitter activity using the liquid scintillator counter. (in Polish). – published in conference proceedings.

1994 A seminar of the Świerk Centre on the "Liquid-scintillation counting technique used for radionuclide activity measurements", Świerk

2001 National Symposium on "Radiochemistry and nuclear chemistry", Kazimierz Dolny – Chyliński A., Broda R., Radoszewski T. National standard of the radionuclides activity unit and related standards (in Polish).

2010 XIII Meeting of SIOR, Dymaczewo Nowe – Broda R. Certification of laboratories with respect to the standardisation of surface contamination detectors. (in Polish).

2011 XIV Meeting of Association of Radiation Protection Inspectors (SIOR), Mikorzyn

– Broda R. A national standard of radionuclide activity units of measure and the system for their submission. (in Polish).

C) Posters at international conferences:

2007 16th ICRM Conference on the “Radionuclide metrology and its applications”, ICRM 2007, Cape Town, South Africa – Broda R. Some remarks on photons statistics in the LS-counter. - published in *Appl. Radiat. Isot.*

2009 17th ICRM Conference on the “Radionuclide metrology and its applications”, ICRM 2009, Bratislava, Slovak Republic – Broda R., Dziel T., Muklanowicz A., Listkowska L., Pieńkowski Ł., Patocka A., Kołakowska E. Comparison of $^{99}Tc^{m}$ and ^{131}I in Polish hospitals, 2007. - published in *Appl. Radiat. Isot.*

D) Posters at national conferences:

2008 XI Symposium of the Polish Society of Nuclear Medicine, Gliwice – Broda R., Dziel T., Muklanowicz A., Listkowska L., Pieńkowski Ł., Patocka A., Kołakowska E. A comparison of $^{99}Tc^{m}$ and ^{131}I measurements at the nuclear medicine units in Poland, 2007. (in Polish).

E) Other international conferences:

1981 International Symposium on the "Methods of low-level counting and spectrometry", West Berlin, Germany – Pochwalski K., Broda R., Radoszewski T., Żelazny P. Standardization of low-level beta-emitter solutions by using the liquid-scintillation triple-to-double coincidence ratio (TDCR) method. – published in conference proceedings.

2005 ICRM conference "Advances in liquid scintillation spectrometry” (LSC 2005), Katowice, Poland – Chairman of the session.

F) Participation in international meetings of specialists:

As the co-author of the works related to the basics of the TDCR method as well as the calculation model of detection efficiency of the LS-counters [Pochwalski et al., 1988; Broda et al., 1988], in 1993, I was invited by dr. T. Quinn, the director of the BIPM, to participate in the works of the Comité Consultatif pour les Étalons de Mesure des Rayonnements Ionisants (CCEMRI), Section II (Radionuclides Measurements). I attended 10 CCEMRI(II) meetings and a series of CCRI(II) meetings, which were working discussions between specialists from dozens of countries on the key comparisons of the radionuclide activity schedule, results of intercomparisons and their uncertainties, functioning the international reference system (SIR), the plan of future Working Groups activity and the reports on the member laboratories activity. I presented a report on the LWR activity and our publications at each meeting of the Consulting Committee. I participated in the ICRM Working Group for Liquid Scintillators activity. I presented the lecture on the analysis of the CIEMAT/NIST method application for the ^{55}Fe activity determination using the ^{3}H as a tracer. I developed counting codes TDCRB-02 (Poisson statistics) and TDCRB-2p (Polya statistics) and their detailed descriptions, which were published on the Working Group's website. I participated in the CCRI(II) Working Group for Extending SIR activity. I presented arguments in discussion of which of the two methods, i.e. TDCR or CIEMAT/NIST, should be implemented in the international reference system extended to include the β-emitters measurements (ESIR).

1993 12th Meeting of Section II of CCEMRI, BIPM, Sèvres, France

1995 13th Meeting of Section II of CCEMRI, BIPM, Sèvres, France

1997 14th Meeting of Section II of CCEMRI, BIPM, Sèvres, France

2000 EUROMET Contact Person Meeting - RAD, Oseras, Norwegia.

I presented the measurement competence and calibration possibilities of the LWR in Świerk and discussed principles of creating the bases for the data storing at the meeting.

2001 16th Meeting of Section II of CCRI, BIPM, Sèvres, France

2002 EUROMET Workshop „Metrology in the European Research Area" (MERA), Rotterdam, Holandia

2003 17th Meeting of Section II of CCRI, BIPM, Sèvres, France, (May 28-30)

2004 BIPM Workshop on CCRI(II) activity comparisons, BIPM, Sèvres, France

2005 18th Meeting of Section II of CCRI, BIPM, Sèvres, France

I presented the preliminary results of the international comparison of activity measurements by means of the TDCR method of ^{63}Ni sources [Razdolescu et al., 2006] at the meeting. I was the mover of the intercomparison with participation of LNHB in France, IFIN-HH in Romania, NAC in South Africa and LWR. The sources were prepared in the Ultima Gold® scintillator, measured in LNHB, and then sent to other laboratories for comparisons. I discussed the comparison results during my visit in Saclay. I collected and analyzed the measurement results and proved existence of differences resulting from various assumptions adopted in the model of the counting efficiency calculation.

2007 19th Meeting of Section II of CCRI, BIPM, Sèvres, France

2008 BIPM Workshop 2 on CCRI(II) activity uncertainties and comparisons, BIPM, Sèvres, France

2009 20th Meeting of Section II of CCRI, BIPM, Sèvres, France

2011 21th Meeting of Section II of CCRI, BIPM, Sèvres, France

2012 Meeting of the ICRM Liquid Scintillation Counting Working Group, Braunschweig, Germany

I presented the lecture on the method of the ionization quenching function determination. I pointed out to the inconsistencies between the adopted assumptions and put forth the hypothesis that introduction of an additional function describing the electron energy loss in the liquid scintillator for energies lower than 1 keV is necessary.

2013 22e Spotkanie Sekcji II CCRI, BIPM, Sèvres, Francja

5.5 Participation in projects:

1991-93 American-Polish Maria Skłodowska-Curie Joint Foundation II Project No. PAA/NIST-91-54 on the "Development of radioactive standard reference materials by liquid-scintillation counting techniques", (in collaboration with the National Institute of Standards and Technology, NIST, Gaithersburg, USA) – performer.

I participated in the measurements of the activity of pure β-emitters measurements using the triple counter. I analysed the elaborated calculation model of the triple counter detection efficiency. [Pochwalski et al. 1988; Broda et al., 1988] and compared the calculation and the experimental results. I modified the applied computing codes.

1994-96 EUROMET Project No. 297 on the "Comparison of activity measurement methods in liquid-scintillation counting and application to the standardization of ^{63}Ni and ^{55}Fe" – main Polish performer.

I introduced the modifications to the ^{63}Ni and ^{55}Fe counting efficiency calculation model and harmonized the computing code algorithm with the LPRI, France. I performed the measurements of a set of sources by means of the TDCR method changing the detection efficiency by various methods and compared them with the CIEMAT/NIST method results. I determined the ionization quenching parameter value for the Ultima Gold® scintillator. The results of the project were published [Cassette et al., 1998].

1995-97 French-Polish Scientific and Technological Cooperation Joint Project No. 194 (212) on the "Development of radioactive standard sources by use of the liquid scintillation technique", (cooperation with LPRI, Saclay, France) – co-ordinator and main Polish performer.

I modified the model of the ^{139}Ce counting efficiency calculation taking into account the Compton photons interaction with the liquid scintillator effects. I measured the set of ^{139}Ce sources by means of the TDCR method and compared it with the results obtained in LPRI [4]. The analysis of the results obtained in both laboratories results enabled us to identify small programming errors and to harmonize the counting codes. I performed the measurements testing of the ^{63}Ni and ^{55}Fe sources stability, which enabled optimization of the scintillation cocktail composition. I performed the measurements of the ^{3}H set of sources within the framework of the international comparison and determined the activities by means of the developed computing code [Broda, 1989]. The analysis of the measurement results enabled us to elucidate the LWR and LPRI results discrepancy.

1998-2000 French-Polish Joint Project POLONIUM No. 98228 on the "Elaboration of a method of activity measurements of α-radionuclides in the presence of β- and EC-emitters in the liquid samples", (cooperation with LPRI, Saclay, France) – Polish co-ordinator and performer.

I applied the TDCR method in the measurements of the radioactive concentration of β-emitters solutions used in the project. I performed the analysis of the model of calculation of the counting efficiency of low-energy β-emitters and demonstrated its limitation for low numbers of the observed scintillation photons. I calculated the ^{3}H and ^{63}Ni activity when the detection efficiency changed in various ways and participated in the joint analysis of the measurement results and preparation of the publication [6].

2001-2002 French-Polish Joint Project POLONIUM 2002 on the "Development the standardization methods of radionuclides used in nuclear medicine by the liquid scintillation technique", (co-operation with LNHB, Saclay, France) – Polish co-ordinator and performer.

In co-operation with the LNHB laboratory, I analyzed the experiments for the stopping power in toluene determination. I used the conclusions in the model of calculation of the detection efficiency of the LS-counters and developed improved counting code TDCRB-02. I performed measurements of a set of sources prepared in various scintillation cocktails with various chemical composition and determined the value of the ionization quenching parameter [Broda et al., 2002].

2011- EURAMET Joint Research Project ENV09 MetroRWM "Metrology for radioactive waste management", (13 JRP partners; coordinator: CMI, Czech Republic) – performer.

I participated in the analysis of the way of preparation of the radioactive waste standards containing ^{137}Cs, ^{60}Co, ^{57}Co, ^{241}Am and ^{90}Sr and the methods of their measurement.

2011- EURAMET Joint Research Project IND04 MetroMetal "Ionizing radiation metrology for the metallurgical industry", (14 JRP partners; coordinator: CIEMAT Spain) – performer.

I participated in analysis the way of preparation of the cast steel, slag and fume dust radioactive standards containing ^{137}Cs, ^{60}Co and ^{226}Ra and the methods of their measurement.

5.6 Didactic, popularization and organizational activity:

RC POLATOM in Świerk is a research and development unit performing research related to the application of the isotopes applications in science, medicine, industry and environment protection. The radiopharmaceuticals and various kinds of radioactive sources and solutions are manufactured in the Centre. RC POLATOM does not carry out the didactic activity in the form of lectures for students. My didactic activity was to take care of two young physicists, student assistants, in LWR (2005, 2012). I trained them in the field of measurement methods applied in the laboratory, ionization radiation detectors and the liquid scintillation technique. The training also comprised performing the radionuclide activity measurements by means the TDCR method, interpretation of the results and evaluation of the measurement uncertainty. I was responsible for two International Atomic Energy Agency (IAEA) student assistants during one-month trainings and students during vacation practice in LWR. I explained problems of the radionuclide metrology and the measurement methods applied in LWR to students groups and specialists visiting OR POLATOM. I worked as a teacher of physics in a secondary school in Warsaw (1972-75).

I presented the results of my work in the field of the radionuclide metrology at RC POLATOM annual seminars. I delivered a lecture on "The LSC technique applied to the radionuclides activity measurement" during the general seminar of the Świerk Centre (1994). Within the framework of the popularization activity, I had a lecture at the Central Office of Measures on the history and development of the radionuclide metrology at the LWR [Broda and Pochwalski, 2011]. I also published 4 articles and I had 2 lectures at the national conferences on the National Standard of Radionuclides Activity Unit existing at the RC POLATOM and its significance in the Metrological System in Poland.

My organizational activity was related to the control the Laboratory of Activity and Radionuclidic Purity Measurement (1977-95) and then the Metrological Laboratory of Radioactive Materials (1996-2002) and LWR (2005-6). I became the Head of the Laboratory when LWR was formed after the RC POLATOM reorganization. I developed an organizational idea of the laboratory and the scope of its activity. One of the outcomes of my activity was the establishment of the National Standard of Radionuclides Activity Unit at RC POLATOM (1999), implementation of the Quality Assurance System and LWR's obtaining the accreditation in 2008. I am the administrator of the national standard. I designed and participated in the launch of a new coincidence LS-counter (TDKG) containing the TDCR counter in the beta channel and the scintillation counter with NaI(Tl) crystal in the gamma channel. Following the test measurements, the TDKG counter will replace some obsolete measurement systems in the national standard.

5.7 Co-operation with institutions, organizations, and scientific societies

A) Scholarships and research contracts in foreign centres:

1987-88 International Atomic Energy Agency Fellowship, LPRI, (12 months)

The fellowship research program concerned the radionuclides activity determination by means of the liquid scintillation technique. I performed the low-energy ^{3}H i ^{55}Fe radionuclides activity measurements using the triple coincidence counter and the TDCR method. I launched the CASIC program controlling the counter and the FORTRAN program for the calculation of the detection efficiency. I have measured the counter characteristics. I investigated the detection efficiency changes using the LED-diode simulated the scintillation source with the PMTs were defocalized. I indicated the necessity to control the PMTs differences when the detection efficiency is changing.

1988 Commissariat a l'Energie Atomique Fellowship, LPRI, Saclay, France, (3 months)

I continued the works conducted during the previous IAEA fellowship.

1999 Contract on the "Standardization of beta emitting nuclides", Brain Pool Program, Korea Research Institute of Standards and Science, Taejon, Republic of Korea (12 months)

I improved the structure of the TDCR counter, proposed a new optical chamber and electronics modules modification as well as performed pure β-emitters activity test measurements. I carried out training for the laboratory workers concerning the basis and principles of the use of the TDCR method as well as on the assumptions adopted in the detection efficiency of LS-counters calculation model. I presented the lecture on the liquid scintillation technique used for the radionuclides activity determination at the conference of the Korean Physical Society.

2002 Contract nt. "Assembling and testing a new TDCR system at NIST", NIST, Gaithersburg, Md, USA (6 weeks)

I launched and tested the triple counter constructed in NIST. I built a new optical chamber and assembled the counter components. I measured the anode characteristics and set the working conditions. I performed the ^{3}H, ^{63}Ni and ^{90}Sr activity measurements by means of the TDCR method. I adjusted the computing codes to the experimental data format and performed calculation using various photons statistical distribution. I delivered a lecture at NIST on "Pure beta emitter activity measurements using the TDCR counter with the triple detector". The results of the work were published [Zimmerman et al., 2004].

2003 Contract on the "Standardising of radionuclides by the TDCR method", European Commission Centre of Excellence IDRANAP, IFIN-HH, Bucharest, Romania (6 weeks)

I improved the functioning of the triple counter constructed in the IFIN-HH. I developed the computing code for the TDCR method and adapted it to the experimental data format. I corrected the discriminator level of the detection system setting. I elaborated the measurement procedure and performed the pure β-emitters ^{3}H and ^{14}C test measurements.

2004 Contract on the "Improving of the TDCR system", European Commission Centre of Excellence Interdisciplinary Research and Applications Based on Nuclear and Atomic Physics (IDRANAP), Horia Hulubei National Institute for Physics and Nuclear

Engineering (IFIN-HH), Bucharest, Romania (1 week)

Continuing the work performed during the contract in 2003, I measured the triple counter anode characteristics and selected the working point. I modified the computing code which allowed me to take into account the PMTs asymmetry and performed the test measurements of the ^{3}H and ^{14}C activity changing the detection efficiency and using optical filters and PMTs defocalization. 1 % consistency of the LWR and LNHB results was obtained.

B) Activity in the Polish metrological system:

1983-2013 Standardization of radionuclides activity.

For over 30 years, I performed the standardization of radioactive solutions and sources produced in the Radioisotope Centre in Świerk and in the Metrological Laboratory, which is the unit controlling the radionuclides activity. I used the relative measurement methods of ionization chambers and scintillation counters with the NaI(Tl) crystals and absolute measurement methods such as 4π(LS)-γ coincidence and anticoincidence, TDCR and the X-γ coincidence methods. I was the head and the performer of over 30 research and development tasks related to the development of the above measurement methods. The results of all the tasks were implemented by the laboratory.

1996-2013 International comparisons of measurement.

The Metrological Laboratory in Radioisotope Centre in Świerk confirmed its metrological competence by participating in dozens international comparisons of radionuclides activity measurements. For over 30 years, I participated in 15 international comparisons of activity measurement of ^{3}H, ^{14}C, ^{32}P, ^{54}Mn, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{89}Sr, ^{90}Sr+^{90}Y, ^{99}Tc, ^{125}I, ^{192}Ir, ^{204}Tl, ^{238}Pu, and ^{241}Am as the performer or the main performer using the 4π(LS)-γ coincidence and anticoincidence and the TDCR methods.

1996-2011 Laboratory Quality System.

I was responsible for the implementation and improvement the Quality System in the Metrological Laboratory of Radioactive Materials, RC, Świerk. Compliance with the system has been the laboratory's duty since the National Standard was established (1999) and kept by the LWR. As the head of the Laboratory, I prepared documents and reviewed the Quality System each year before the GUM audit. I was the co-author of the documentation prepared for the LWR accreditation (2008).

1999 National Standard Administrator.

I was the main author of the specification for establishing the National Standard Unit of Radionuclides Activity in Poland drawn up in accordance with the requirements laid down by the President of the Central Office of Measures (GUM). The standard was established in the Radioisotope Centre in Świerk of the Metrological Laboratory of Radioactive Materials which I managed. I became the National Standard Administrator. Obtaining the National Standard certificate enabled the LWR's standardization of the radioactive sources and solutions and calibration of the activity calibrators for the users in Poland.

2005-2013 PCA technical auditor.

On behalf of the Polish Accreditation Center (PCA), I have participated in 40 audits of some laboratories of radiation dosimetry in Poland. I evaluated technical competence of laboratories for standardization in the accreditation field, laboratory staff competences, credibility of the results demonstrated and verified if the Laboratory Quality System was in accordance with the PN-EN ISO/IEC 17025:2005 standard.

C) Activity in national and international professional societies:

1992- Polish Physical Society - member

1995- Section II of CCRI (former: CCEMRI) - delegate, Working Group on Extension of the SIR, BIPM, Sèvres, France

I participated in discussions about the extension the international reference system SIR to include the measurements of the pure β-emitters and the development of the reference liquid scintillator. I explained the experimental and calculation problems in application of the TDCR method.

1995-2005 Scientific Council of the Radioisotope Centre - member, Świerk.

I participated in the Scientific Council of the Radioisotope Centre activity and, among other things, in discussions about the Centre reorganization. I participated in analysing the economical results of the Centre activity and in preparation the reports for the Scientific Council as a member of the Economic Commission.

1997- ICRM – member of the Executive Board, Working Group on Liquid – Scintillation Counting

I participate in discussing the current activity of the ICRM and fixing the schedule of its work. I participate in the Working Group specialists meetings. I present lectures at the periodical ICRM conferences organized each two years.

2005 - Polish Centre for Accreditation – technical auditor, Warsaw

I was delegated by the Association Française de Normalisation AFNOR to participate in the accreditation audit in GUM (2005). I obtained the licence of the technical expert of PCA. I participate in audits of laboratories for dosimetry of ionizing radiation in Poland.

2011- Committee for Metrology and Scientific Instrumentation of the Polish Academy of Science – member, Wroclaw, Poland

In connection with my activity in the field of radionuclide metrology and administrating of the National Standard in the Radioisotope Centre POLATOM being the Designated Institute (DI), I was invited by prof. J. Mroczka, the president of the Committee for Metrology and Scientific Instrumentation of the Polish Academy of Science (KMiAN PAN) to join this Committee and appointed as a member. I participated in the preparation of the data for the report on the status and perspectives of development of the metrology as a scientific inter-discipline, which was edited by prof. J. Gajda and published in the KMiAN PAN activity guide-book (2014). I prepared a forecast related to the perspective of the ionization radiation measurements development until 2020.

Warsaw, 30.05.2014

Ryszard Broda